Statistical and Low Temperature Physics (PHYS393)

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9. The magnetic refrigerator

Sections

We have seen that using dilution of helium-3, we could reach temperatures in the milliKelvin range.

If we want to go below that, a completely new technology is needed. This can be achieved using magnetic cooling. Historically, magnetic cooling has developed in 2 stages.

When it was first proposed in the 1920s, paramagnetic salts was used for cooling. Today, this method can each down to milliKelvin temperatures. The use of paramagnetic salt has now been largely replaced by the dilution refrigerator.

In the 1950s, the use of the magnetic moment of nuclei in metals was started. This method can now reach microKelvin temperatures.

Whether it is magnetic moments of electrons in salts or nuclei in metals, the principle is the same.

9.1 The Principle of Magnetic Cooling

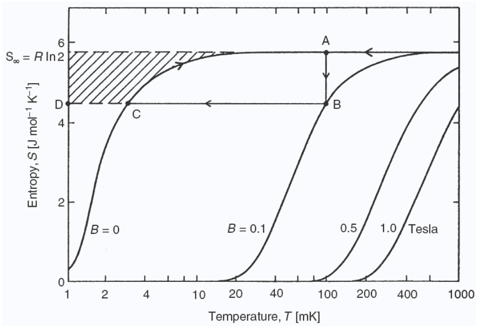
Consider a paramagnetic salt. This salt contains ions with magnetic moments coming from their electrons.

At mK temperatures, the magnetic disorder entropy (about 1 J/mol) is large compared to all other entropies, such as lattice and coduction electron entropies, which may be neglected.

We have previously looked at the properties of a paramagnetic salt. The entropy would be a function of the magnetic field applied. We shall look at the derivation of the entropy formula later.

First, we look at the known behaviour of the entropy and see how this can help us to understand magnetic cooling.

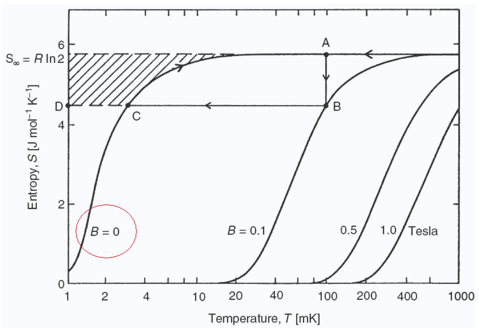
This graph show the entropy against temperature for a commonly used paramagnetic salt.



S.A.J. Wiegers, P.E.Wolf, L. Puech: Physica B 165 & 166, 165 (1990)

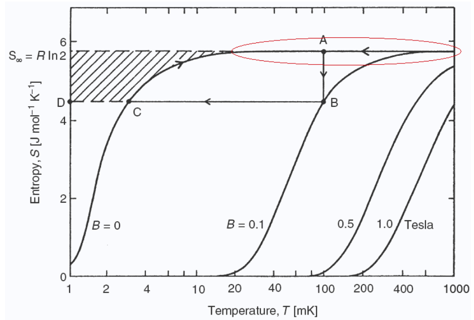
We start by familiarising ourselves with the various features of the graph.

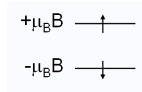
The symbol B is the magnetic field. We start with the graph for zero or low magnetic field.



The Principle of Magnetic Cooling

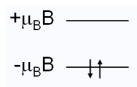
At high temperature, the entropy approaches a constant because it becomes equally likely to be at any of the magnetic energy levels.

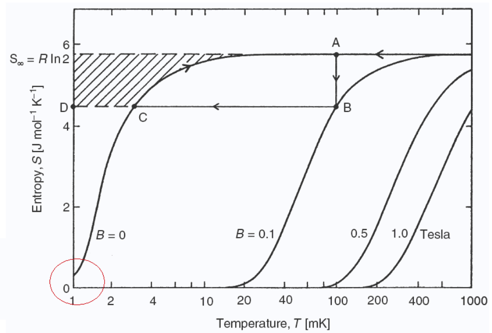




The Principle of Magnetic Cooling

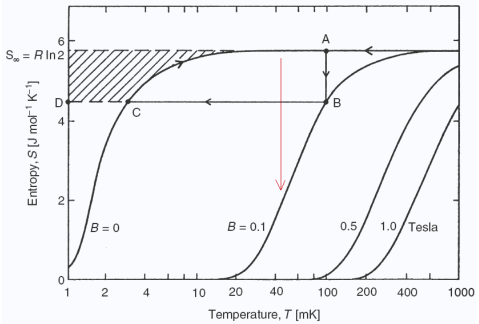
At low temperature, the entropy goes to zero because all particles fall to the lowest level.





The Principle of Magnetic Cooling

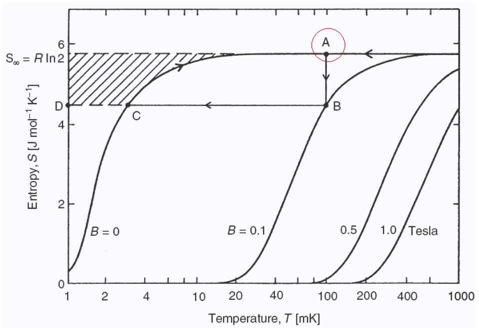
Next, suppose the magnetic field is increased, say from 0 T to 0.1 T. The spacing between energy level would increase.



Then it becomes more likely for a particle to be at the lower level. So the entropy would fall.

The Principle of Magnetic Cooling

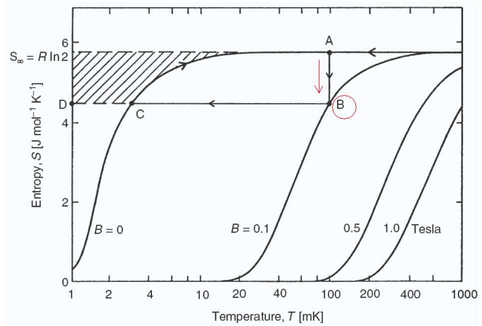
Point A: To understand how to use the magnetic property for cooling, suppose we start with a temperature at point A, and with a low magnetic field..



The salt is placed in contact with a precooling bath. This can either be a helium bath, or a dilution refrigerator.

The Principle of Magnetic Cooling

Point B: A magnetic field is then applied. This is done isothermally. The entropy falls to point B at constant temperature.

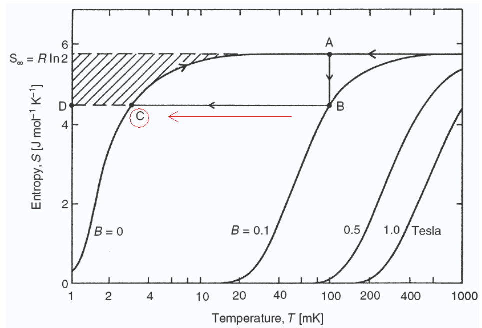


This process performs magnetic "work" on the salt, which is converted to heat (like compressing a gas). This heat would be absorbed by the precooling bath.

The Principle of Magnetic Cooling

The salt is then thermally isolated from the precooling bath (e.g. by using a heat switch).

Point C: Demagnetisation now takes place adiabatically (so entropy is constant). The magnetic field is reduced to a very small value. The temperature falls to C.

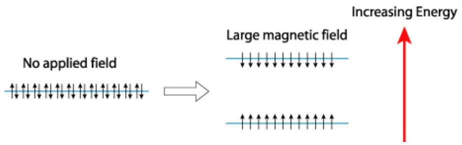


How does it work?

We have seen how the cooling takes place using thermodynamics. Let us now see how this takes place physically.

The ions in the salt have magnetic dipole moments. Normally, half of the dipoles are spin up, and the other half are spin down.

If a strong magnetic field is applied, the energy levels will split into two.

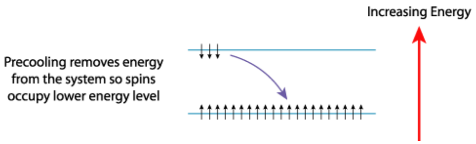


Dipoles in the direction of the field will have lower energy, and dipoles in the opposite direction higher energy.

Precooling

Remember that a helium bath or a dilution refrigerator is cooling the salt at the same time.

This would remove energy from the higher energy atoms, so that they fall into the lower energy state.

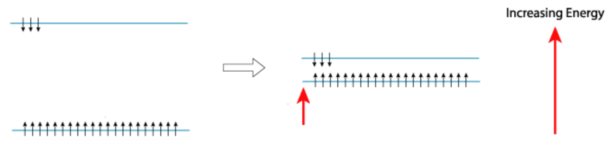


This is the "precooling."

Then, using a heat switch, the salt can be thermally disconnected from the precooling bath.

The magnetic field is now slowly reduced.

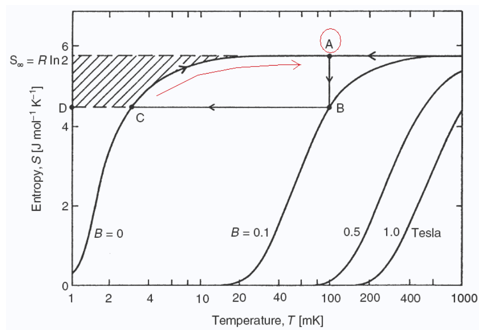
The lower energy level, which contains most of the atoms, is then forced to increase in energy.



This energy has to come from the surrounding. So the salt cools down.

The Principle of Magnetic Cooling

Point C: After some time, the salt would warm up because of heat leak from the surroundings, since the insulation is not perfect. The temperture returns to point A along the curve from C.



The temperature cannot be maintained, unlike the dilution refrigerator. This is called a "one-shot" technique.

9.2 Thermodynamics of Magnetic Cooling

Thermodynamics of Magnetic Cooling

With the help of the entropy graph, we can calculate the heat Q and temperature T in magnetic cooling.

We can divide the cycle into the following stages:

1. Isothermal magnetisation: We shall find the heat given out by the salt.

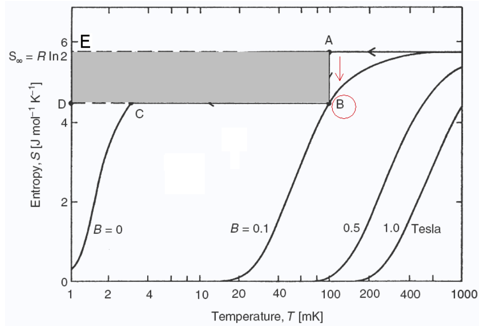
2. Adiabatic demagnetisation: We shall find the lowest temperature reached.

3. Warming up: We shall determine the cooling power.

Isothermal magnetisation takes place from A to B on the graph at the start. Since $dQ = TdS$, the heat given out is

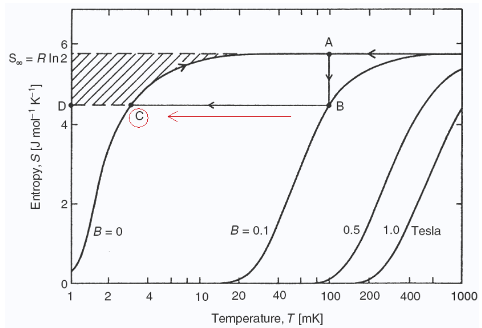
$$Q = \int_B^A TdS.$$

This is just the area of the rectangle ABDE.



The heat released is usually a few J/mol of the refrigerant (the salt), so it can easily be absorbed by an evaporating helium bath or a dilution refrigerator.

Adiabatic demagnetisation takes place from B to C on the graph.



Later on, we shall derive the formula for magnetic entropy. In that formula, we shall see that the entropy is a function of B/T.

For now, a quick way to understand this is to look at the Boltzmann distribution, $\exp(-\mu_B B/k_B T)$. This is indeed a function of B/T.

Since the entropy should depend on $\exp(-\mu_B B/k_B T)$, we would expect the entropy to be a function of B/T as well.

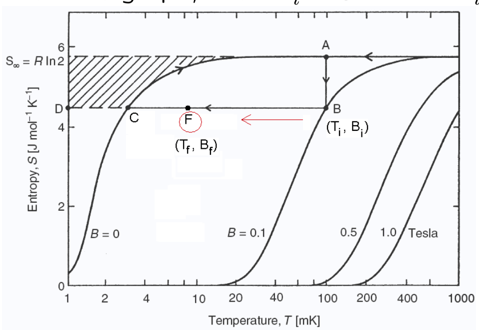
This gives us a quick way to find the coldest temperature reached in this demagnetisation step.

From the graph, we see that the entropy is a simple function of T and B. If T increases, entropy increases. If B increases, entropy decreases.

In the adiabatic process, the entropy is a constant. Since it is a function of B/T, then B/T must also be constant. This is just the equation we need:

$$\frac{B}{T} = \text{constant}$$

Point B: Suppose we start at temperature T_i and field B_i. If this is point B on the graph, then $B_i = 0.1T$ and $T_i = 100mK$



Point F: We then reduce the field to a smaller value B_f. Let T_f be the new temperature.

The equation B/T = constant means that

$$\frac{B_f}{T_f} = \frac{B_i}{T_i}$$

The new temperature is

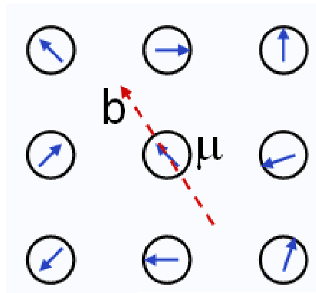
$$T_f = \frac{T_i}{B_i} B_f$$

Clearly, we can make T_f very small by reducing the magnetic field B_f to a very small value.

But what if we reduce the magnetic field B_f to zero? Surely the temperature T_f would not go to zero. Something must happen to limit the lowest temperature that we can reach.

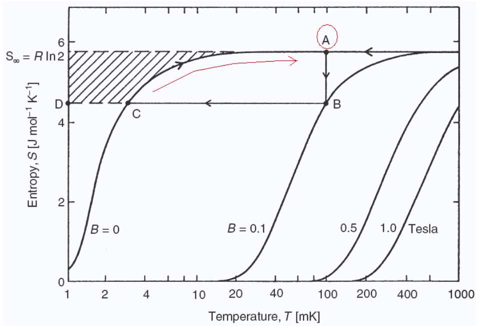
Indeed, this is the case. When the temperature is sufficiently low, the effects of the magnetic fields from neighbouring ions of the salt become important.

When the temperature is sufficiently low, the mutual interaction would tend to align all magnetic dipoles in the same direction. When this happens, the entropy falls to zero, and the magnetic cooling would stop.



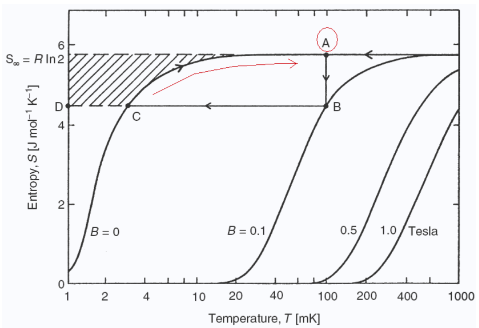
So the mutual interaction limits the lowest temperature that can be achieved using this method. At very low temperatures, the equation would have to be modified to take into account this mutual interaction. We shall come back to this.

Point C: We can now see that the graph is misleading. We do not actually demagnetise to a field at point C, which is zero. Rather, we would demagnetise to a very small field, close to C.



To make things simple, we shall still refer to C for the end point of the demagnetisation. Then warming up starts.

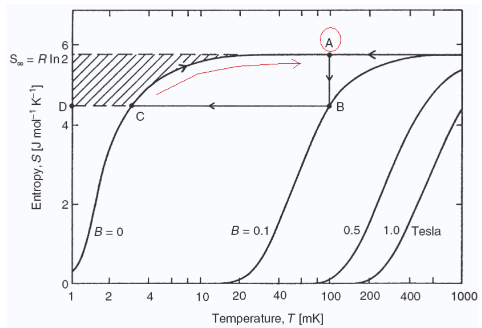
Remember that the salt is thermally isolated during the demagnetisation. After reaching the lowest temperature at C, the salt remains isolated. We hope that it would stay cold for as long as possible.



But because insulation is not perfect, the salt starts warming up slowly. Since the magnetic field B is fixed, the temperature and entropy would follow the curve and eventually reach the starting temperature at A.

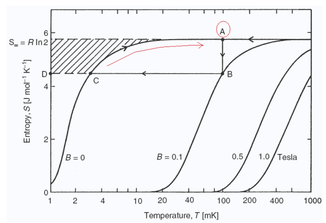
As it warms up, the heat absorbed by the salt can be obtained from the entropy using the same formula as before

$$Q = \int_C^A T dS$$



We need to integrate along the curve from C to A. So the heat absorbed is given by the shaded region on the graph.

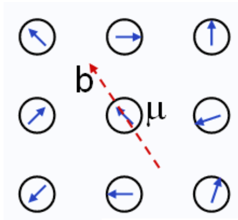
The heat absorbed in warming up also gives the cooling power. If the salt can absorb more of the heat that leaks in through the insulation, then it would be able to remain cold for a longer period of time.



Note that this is a different definition from before. For the dilution refrigerator, the cooling power $\dot{Q}$ is the rate at which heat is absorbed.

The cooling power Q for the magnetic refrigerator is the total heat absorbed.

We have seen that the interaction between magnetic dipoles of the ions in the salt sets a lower limit to the temperature that can be reached by demagnetisation.



This also means that the formula for the lowest temperature

$$T_f = \frac{T_i}{B_i} B_f$$

would not be accurate at very low temperatures. It can be modified to the following form:

$$T_f = \frac{T_i}{B_i}\sqrt{B_f^2 + b^2}$$

Lets try and understand this formula physically.

$$T_f = \frac{T_i}{B_i}\sqrt{B_f^2 + b^2}$$

We see that when B_f is reduced to zero,

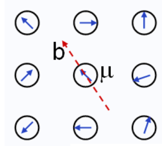
$$T_f = \frac{T_i}{B_i} b.$$

If we compare this with the original form of

$$T_f = \frac{T_i}{B_i} B_f$$

we see that b corresponds to B_f. This makes sense if we think of b as the field that remains after the applied magnetic field has been reduced to zero.

When the applied magnetic field is reduced to zero, there is indeed a remaining field. That would be the resultant field from the neighbouring magnetic dipoles.



We are looking at a temperature T_c at which the effect of this interaction becomes important. This means that k_BT_c is comparable to the interaction energy

$$\varepsilon_d = \mu b.$$

T_c is called the ordering temperature. It is the temperature below which the neighbouring fields become strong enough to align the dipoles. We may define

$$k_BT_c = \mu b.$$

9.4 Magnetic disorder entropy

Previously, in the lectures on Paramagnets, we have seen that the entropy can be derived from the partition function. We look at this in more details now.

The partition function is

$$Z = \sum_{-J}^{+J} e^{-\varepsilon_m/k_BT}$$

where

$$\varepsilon_m = \mu_B g m B$$

and g is the Landé factor.

This assumes that there is no interaction between the magnetic dipoles. The only relevant energy comes from the applied field B acting on the individual dipole $g\mu_B$.

The entropy can be derived from the partition function using

$$S = Nk_B \ln Z + Nk_BT\frac{\partial \ln Z}{\partial T}$$

where N is the number of particles. We shall derive the entropy for 1 mole of the salt, so N would be Avogadro's number, and $Nk_B = R$, the ideal gas constant.

Note that the equation can also be written in this form:

$$S = R\frac{\partial (T \ln Z)}{\partial T}$$

For convenience, let $x = \mu_B g B/k_BT$. The partition function is then

$$Z = \sum_{-J}^{+J} e^{-mx} = \frac{\sinh[(J+1/2)x]}{\sinh(x/2)}$$

Substituting the partition function

$$Z = \frac{\sinh[(J+1/2)x]}{\sinh(x/2)}$$

into the formula for the entropy

$$S = R\frac{\partial(T\ln Z)}{\partial T}$$

gives

$$\begin{aligned}\frac{S}{R} &= \left(\frac{x}{2}\right)\left\{\coth\left(\frac{x}{2}\right) - (2J+1)\coth\left[\left(\frac{x(2J+1)}{2}\right)\right]\right\}\\ &+ \ln\left[\frac{\sinh[x(2J+1)/2]}{\sinh(x/2)}\right]\end{aligned}$$

The small x expansion of this formula is of particular interest. Recall that $x = \mu_B gB/k_B T$. Small x would correspond to high T. This is a regime that would be useful when the nuclear magnetic moment is used for cooling.

To derive the small x expansion for this formula,

$$\begin{aligned}\frac{S}{R} &= \left(\frac{x}{2}\right)\left\{\coth\left(\frac{x}{2}\right) - (2J+1)\coth\left[\left(\frac{x(2J+1)}{2}\right)\right]\right\}\\ &+ \ln\left[\frac{\sinh[x(2J+1)/2]}{\sinh(x/2)}\right]\end{aligned}$$

we need the following expansions:

$$\begin{aligned}\sinh x &= x + \frac{1}{6}x^3 + ...\\ x\coth x &= 1 + \frac{1}{3}x^2 + ...\end{aligned}$$

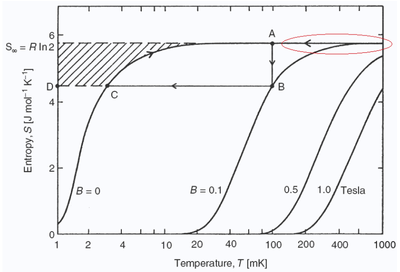
Applying the expansion for $x\coth x$ to the above entropy formula, we obtain

$$\begin{aligned}\frac{S}{R} &= \ln(2J+1) - \frac{1}{6}J(J+1)x^2 + ...\\ S &= R\ln(2J+1) - \frac{1}{6}J(J+1)R\frac{\mu_B^2 g^2 B^2}{k_B^2 T^2} + ...\end{aligned}$$

The expansion of the entropy to second order,

$$S = R\ln(2J+1) - \frac{1}{6}J(J+1)R\frac{\mu_B^2 g^2 B^2}{k_B^2 T^2},$$

would be useful if the nuclear magnetic moment is used for cooling.



The magnetic energy would be much smaller than $k_B T$, so that the temperature is high by comparison. We shall look at this in more details.

9.5 Magnetic Refrigerators

The performance of a magnetic refrigerator is mainly determined by:

- the starting magnetic field and temperature,
- the heat leaks, and
- the paramagnetic salt that is used.

Typical starting conditions are 0.1 to 1 T and 0.1 to 1 K. These are fairly easy to achieve nowadays.

There a few properties of a paramagnetic salt that are desirable:

- low ordering temperature to reach low temperatures,
- large specific heat to absorb more heat before warming up

The following are paramagnetic salts that have been used:

"High"-temperature salts:

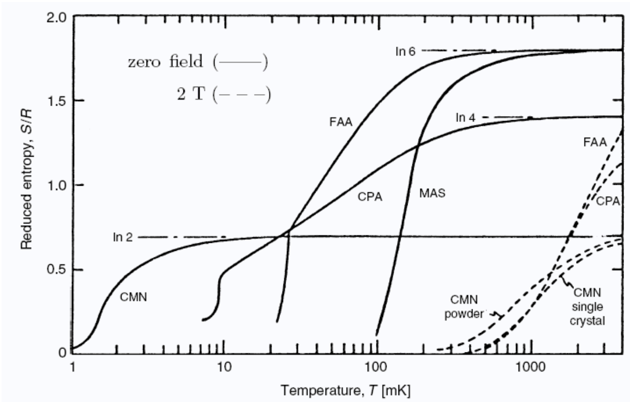
$$\mathrm{MAS}: \mathrm{Mn}^{2+}\mathrm{SO}_4 \cdot (\mathrm{NH}_4)_2\mathrm{SO}_4 \cdot 6\mathrm{H}_2\mathrm{O}; \qquad T_c \simeq 0.17\,\mathrm{K}$$
$$\mathrm{FAA}: \mathrm{Fe}_2^{3+}(\mathrm{SO}_4)_3 \cdot (\mathrm{NH}_4)_2\mathrm{SO}_4 \cdot 24\mathrm{H}_2\mathrm{O}; \qquad T_c \simeq 0.03\,\mathrm{K}$$

"Low"-temperature salts:

$$\mathrm{CPA}: \mathrm{Cr}_2^{3+}(\mathrm{SO}_4)_3 \cdot \mathrm{K}_2\mathrm{SO}_4 \cdot 24\mathrm{H}_2\mathrm{O}; \qquad T_c \simeq 0.01\,\mathrm{K}$$
$$\mathrm{CMN}: 2\mathrm{Ce}^{3+}(\mathrm{NO}_3)_3 \cdot 3\mathrm{Mg}(\mathrm{NO}_3)_2 \cdot 24\mathrm{H}_2\mathrm{O}; \qquad T_c \simeq 0.002\,\mathrm{K}$$

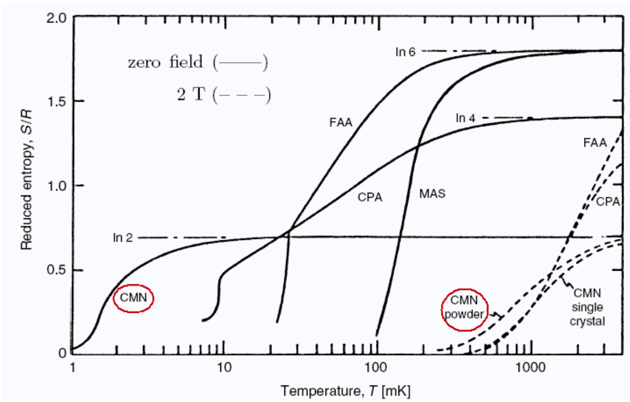
The last one, CMN, has the lowest ordering temperature. This means it can potentially reach the lowest temperature before the interaction between magnetic dipoles become important. CMN has been used extensively and could reach 2 mK.

The follow graphs show the entropies of the actual salts.



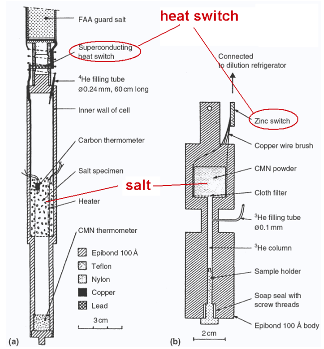
Pobell (2007)

For example, look at the 2 graphs for the salt CMN:
The one to the left is for zero magnetic field.
The one to the right is for 2 T.



They look very similar to the entropy graphs that are shown earlier.

These are examples of actual magnetic refrigerators that have been built.



Pobell (2007)

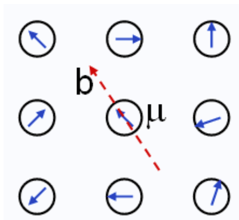
Notice the heat switch near the top. They are usually connected to a dilution refrigerator. The paramagnetic salt refrigerant is in the middle.

Magnetic refrigerators using paramagnetic salts are now largely replaced by the dilution refrigerator, which can reach the same temperatures.

However, they are still useful for small experiments and satellites, where compact refrigerators are required. Examples are in detectors for millimetre wave, X rays and dark matter.

9.6 Nuclear Refrigeration

We have so far looked at the use of the electronic magnetic dipoles for cooling. This is limited to milliKelvin temperatures by the interaction between the electronic dipoles.



It is possible to reach much lower temperatures if we use the nuclear magnetic dipoles. The magnetic dipole moment of the nucleus is much smaller than that of the electron. As a result, the interaction between nuclear dipoles is much weaker.

To get some idea of the relative magnitudes, we look at the unit for electronic dipole moment (Bohr magneton) and the unit for nuclear dipole moment (nuclear magneton):

Bohr magenton, $\mu_B = 9.27 \times 10^{-24}$ J/T
Nuclear magenton, $\mu_n = 5.05 \times 10^{-27}$ J/T

The nuclear magneton is nearly 2000 times smaller. This gives us an idea of how much smaller the nuclear magnetic moment is.

If we use the nuclear magnetic dipole for cooling, we can reach microKelvin temperatures because of the much smaller interaction field. The ordering temperature for the nuclear dipole can be as small as 0.1 μK.

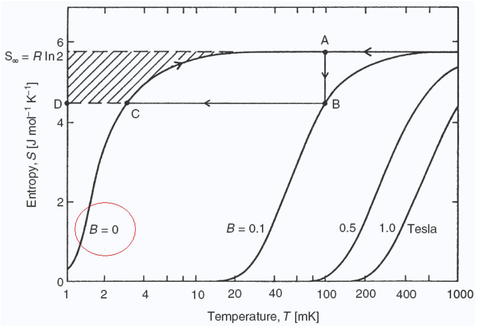
For nuclear cooling, we can use metal as the refrigerant instead of paramagnetic salts. Metal has the advantage of high thermal conductivity.

Although the small nuclear moment offers the potential of reaching much lower temperatures, it also requires much more demanding conditions. The very small moment means that we need very high starting magnetic fields, and very low starting temperatures.

As an example, we look at copper. Copper is a "work horse" of nuclear refrigeration. For copper, we would typically need a starting field of $B_i = 8$ T, and a starting temperature of $T_i = 10$ mK. This is just to reduce the entropy by 9%.

From the earlier explanation on the principle of magnetic cooling, we know that an lower entropy means that a lower temperature can be reached during demagnetisation. It also means a higher cooling power, since more heat can be absorbed during warming up.

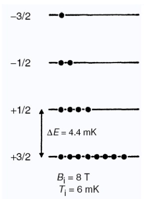
From the entropy graph, we can see that to reduce the entropy further, even higher fields and lower temperatures would be required.



The 8 T field already requires superconducting magnets, and the 10 mK temperature would require a dilution refrigerator.

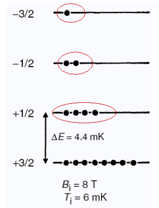
Why would a small magnetic moment require higher starting field and lower starting temperature? Lets try and understand this physically.

Consider the magnetic energy levels of copper. Applying a magnetic field increases the spacing between levels. Because of the small nuclear moment, the spacing would be small even for a high starting field.



Pobell (2007)

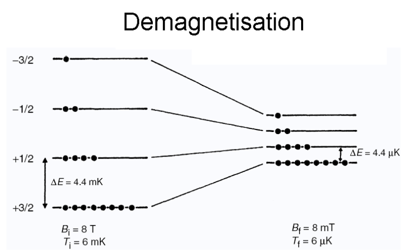
Because the spacing is small, we would get more particles at the higher energy level according to the Boltzmann distribution. This means higher entropy. In order to reduce this, we need a lower starting temperature.



Pobell (2007)

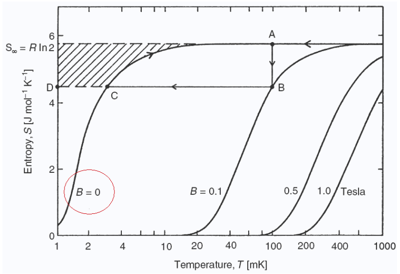
This is difficult. For copper, even at a starting temperature of 10 mK, there is still a substantial fraction of the nuclei at the higher levels. The levels are just too close together because of the small nuclear moment.

Fortunately, because the nuclear moment is small, the interaction field is also small. This means that in the demagnetisation step, it is possible to reduce the temperature to a very small value.



In the case of the copper example, if we reduce the field by 1000 times to 8 mT, the temperature also falls by 1000 times to 6 μK.

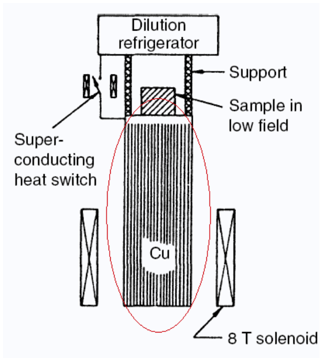
One disadvantage of very low temperatures in magnetic cooling is that the cooling power becomes very small. The cooling power is given by the shaded region in the graph.



The horizontal axis is temperature. So for low temperatures, the horizontal size of the shaded area would also be small. Since nuclear cooling is 1000 times colder than electronic magnetic cooling, the cooling power is also 1000 times smaller.

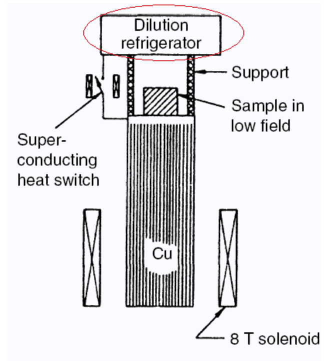
This is a schematic diagram of a nuclear refrigerator. Notice how the main components are connected together.

The refrigerant is a copper block at the centre.

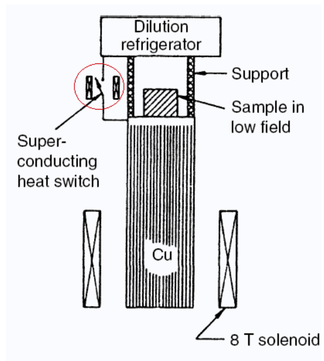


Pobell (2007)

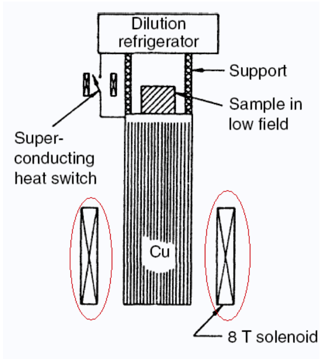
The copper block is connected to the dilution refrigerator. This cools the copper to the starting temperature.



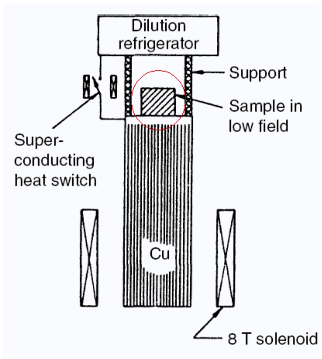
In between the copper block and the dilution refrigerator is a heat switch. This thermally isolates the copper once it reached the starting temperature.



The copper block is surrounded by a solenoid which supplies the magnetic field. This is reduced to a very small value during demagnetisation.



The sample to be refrigerated is in thermal contact with the copper block. The good conductivity of copper makes it easy to cool the sample.



Together with the dilution refrigerator, the whole set up would be surrounded by a vacuum and a helium bath.

The small nuclear moment makes it possible to use a simpler equation for the entropy. Recall that we have previously obtained a second order expansion of the entropy:

$$\frac{S}{R} = \ln(2J+1) - \frac{1}{6}J(J+1)x^2 + \ldots$$

where $x = \mu_B g B / k_B T$. For nuclear cooling, we would replace the Bohr magneton μ_B with the much smaller nuclear magneton μ_n, so x would be small.

So $x = \mu_n g B / k_B T$. The nuclear spin is usually denoted by I instead of J. Substuting these, the nuclear entropy per mole can be written as:

$$S_n = R\ln(2I+1) - \frac{\lambda_n B^2}{2\mu_0 T_n^2}$$

where $\lambda_n = N_A I(I+1)\mu_0 \mu_n^2 g_n^2 / 3k_B$, and g_n is the Landé factor for the nucleus.

$$S_n = R\ln(2I+1) - \frac{\lambda_n B^2}{2\mu_0 T_n^2}$$

In this formula, the temperature is written as T_n. The subscript n emphasises that the cooling takes place among the nuclei. In order for the metal containing these nuclei to refrigerate other samples, the cooling in the nuclei must first be transmitted to the electrons and the lattice of the metal.

Depending on the metal and the conditions, this can take quite long, possibly longer than the time it takes for the nuclei to warm up.

There are also very powerful nuclear refrigerators that cannot be described by the above formula for entropy. These use even higher fields and lower temperatures, so that $x = \mu_n g B / k_B T$ is no longer small. Physically, this means that the magnetic energy $\mu_n g B$ is no longer small compared to the thermal energy $k_B T$.

From the entropy formula,

$$S_n = R\ln(2I+1) - \frac{\lambda_n B^2}{2\mu_0 T_n^2},$$

we can readily obtain the heat capacity. Since the volume of the metal is constant, the energy increase is equal to the heat absorbed:

$$dU = T dS.$$

So the heat capacity is

$$C = \frac{dU}{dT} = T\frac{dS}{dT}.$$

Differentiating the entropy equation above with respect to T, we get heat capacity per mole:

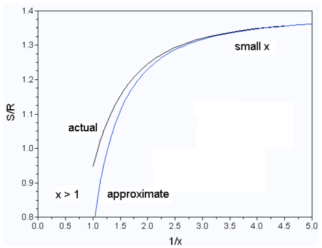
$$C_n = \frac{\lambda_n B^2}{\mu_0 T_n^2}, \qquad \text{(Schottky law)}$$

where $\lambda_n = N_A I(I+1)\mu_0 \mu_n^2 g_n^2 / 3k_B$.

In order to use this approximation for the entropy

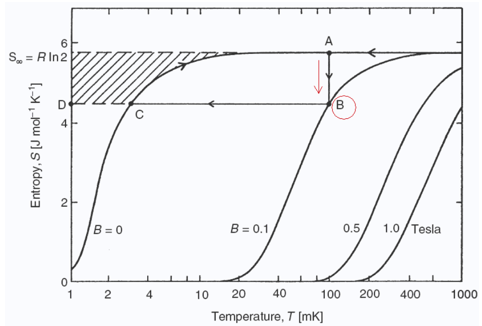
$$\frac{S}{R} = \ln(2J+1) - \frac{1}{6}J(J+1)x^2$$

it would be useful to know the range of x ($= \mu_B g B / k_B T$) for which it is valid. In the following figure, this is plotted against $1/x$, which increases in the same direction as the temperature.



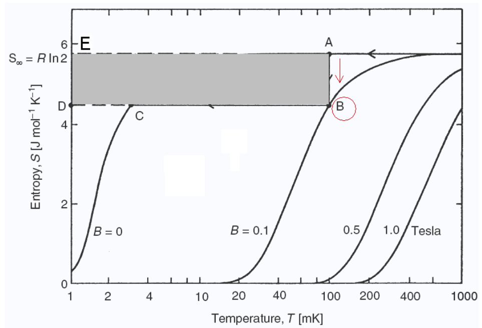
From a graph like this, we can see that the approximation is accurate at small x (to the right of the graph). It would deviate significantly from the correct entropy for $x > 1$ (to the left).

To see how we may apply the approximation for entropy, we look at the heat absorbed during the precooling stage. For nuclear cooling, there are two ways to precool the nuclear moments.



One way is the same as the electronic magnetic cooling using paramagnetic salt - to increase the magnetic field isothermally.

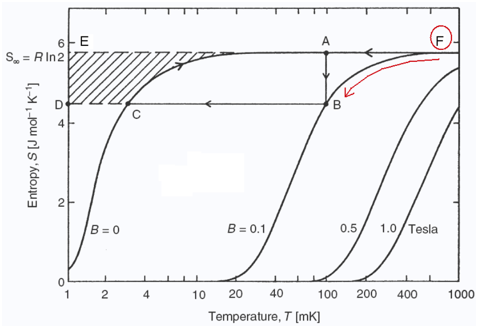
A dilution refrigerator can be used to absorb the heat, Q_A, produced by the magnetisation. This is given by the area ABDE.



Using the approximate formula for entropy, we find:

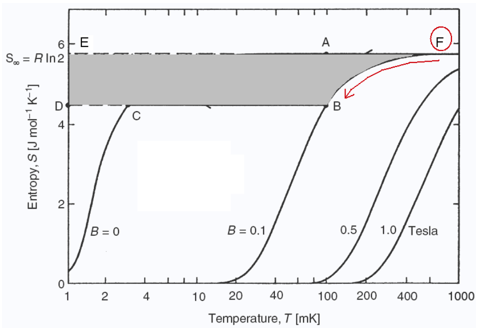
$$Q_a = nT_i \Delta S = -\frac{n\lambda_n B_i^2}{2\mu_0 T_i}$$

The other way to precool is to switch on the field B_i at high temperature, and keep the field fixed. Then the temperature is lowered to the starting value T_i.



For example, instead of point A, we would start at point F, which is at a higher temperature. Since the field is fixed, as we cool down, we would follow the curve until we reach the point B.

The heat of magnetisation would be higher. It is given by the area FBDE.

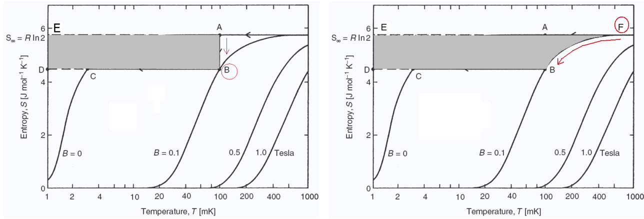


This area can also be obtained by integrating with the approximate formula. The heat given out is:

$$Q_b = nT_i \Delta S = -\frac{n\lambda_n B_i^2}{\mu_0 T_i} = 2Q_a$$

Note that using the second way of precooling, the amount of heat that is given out during magnetisation is doubled.

The heats for the two ways of precooling are given by the shaded areas below. The area on the right may not look twice as big, but remember that the horizontal axis is in log scale.



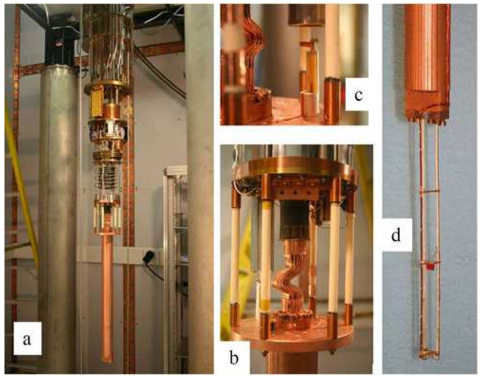
Even so, the second way is often used. The reason is that the dilution refrigerator has a higher cooling power at the higher temperature. This makes up for the cooling time, which may actually be shorter.

This is a dilution refrigerator built by Oxford Instruments. It is used in the Rice University (USA) to study ultracold 2D electron systems.

http://www.ruf.rice.edu/~dulab/ResearchUltraCold.html

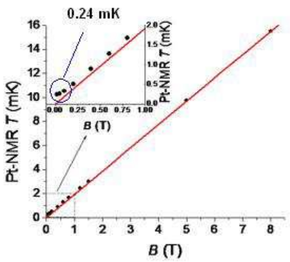
On the left is the assembled system - the blue cylinder is a huge helium dewar. On the right is the dilution refrigerator that is inserted into the dewar.

(a) The nuclear refrigerator is attached to the dilution refrigerator. Notice the large copper block at the botton.
(b) Aluminum heatswitch,
(c) Platinum NMR thermometer,
(d) Sample holder assembly.



http://www.ruf.rice.edu/~dulab/ResearchUltraCold.html

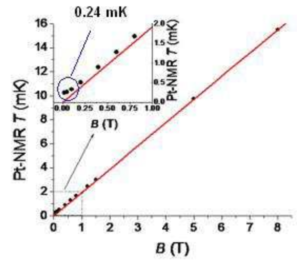
The measured copper temperature versus demagnetization field is shown. The line shows ideal adiabatic relation, $B/T = B_i/T_i$.



http://www.ruf.rice.edu/~dulab/ResearchUltraCold.html

The nuclear stage was precooled in an 8 T field down to 15.5 mK with the dilution unit in 18 hours. After thermally isolating the stage by opening the heatswitch, the copper was demagnetized down to 20 mT.

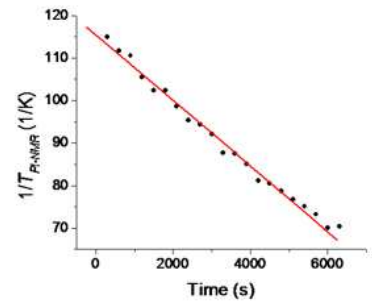
Notice from the inset that the data stops at 0.24 mK.



http://www.ruf.rice.edu/~dulab/ResearchUltraCold.html

This is because Pt-NMR thermometer starts to decouple thermally from the copper-stage below about 0.5 mK. This could be because the platinum becomes superconducting and starts acting like the heat switch. This means that the thermometer would stop working.

The inverse $1/T$ of the temperature is plotted against time. The falling graph means that the temperature is rising with time. This happens when the demagnetisation stops.



http://www.ruf.rice.edu/~dulab/ResearchUltraCold.html

This comes from heat leak. It can be estimated from this that the heat leak is about 21 nW, which is quite typical. This data tell us how much time we have for our experiments before it warms up.

9.7 Exercises

Exercise 1

The high temperature approximation for the magnetic heat capacity of a spin-1/2 salt is given by

$$C_a = Nk_B\left(\frac{\mu_B B}{k_B T}\right)^2 .$$

The exact result is given by

$$C = Nk_B\left(\frac{\mu_B B}{k_B T}\right)^2 \text{sech}^2\left(\frac{\mu_B B}{k_B T}\right).$$

For a field of 1 T, at what temperature does the high temperature approximation for the magnetic heat capacity deviate by 5% from the exact results?

Define

$$x = \frac{\mu_B B}{k_B T}.$$

Rewrite the given equations in terms of x. The high temperature approximation is

$$C_a = N k_B x^2.$$

The exact result is

$$C = N k_B x^2 \mathrm{sech}^2 x.$$

sech$x$ is a decreasing function. When they differ by 5%, the difference is

$$\frac{C - C_a}{C_a} = 0.05.$$

Substituting the above equations, we get

$$1 - \mathrm{sech}^2 x = 0.05.$$

This

$$1 - \mathrm{sech}^2 x = 0.05$$

can be solved to give

$$x = 0.3230$$

or

$$\frac{\mu_B B}{k_B T} = 0.3230.$$

We are given that the field B is 1 T. Substituting this and the constants, we find

$$T = 2.08 \text{ K}.$$

Exercise 2

CMN is a spin-1/2 paramagnetic salt. Calculate the cooling power of one mole of CMN if it is demagnetised from 2 T, 1 K to zero field. Calculate for the same experiment the heat of magnetisation which has to be removed if this salt is magnetised isothermally to 2 T.

[You are given that the magnetic ordering temperature of CMN is 2 mK.]

When cooled to zero field, an interaction field b remains. This is related to the magnetic ordering temperature T_c by

$$\mu_B b = k_B T_c.$$

Substituting the ordering temperature given and the constant, we find

$$b = 0.00298 \text{ T}.$$

This is the minimum field that remains after the applied field is reduced to zero. The final temperature T_f is then given by

$$T_f = \frac{T_i}{B_i} B_f$$

The initial temperature T_i is 1 K, and initial field B_i is 2 T. Taking the interaction fiend b as the final field B_f, we find

$$T_f = 0.00149 \text{ K}.$$

The cooling power is the heat needed to warm up the salt from this temperature. This is given by

$$Q = \int_{T_f}^{\infty} C dT$$

where C is the heat capacity. It is a good approximation to the upper limit to infinity, because the heat capacity falls as $1/T^2$. So Q would reach a limiting value at higher temperature.

Since the heat capacity C is obtained by differentiating the energy U with respect to temperature T, we have

$$Q = \int_{T_f}^{\infty} C dT = U(\infty) - U(T_f).$$

For a spin-1/2 salt, we know from the lectures on paramagnetic salts that the energy is given by

$$U = -N\mu_B B \tanh\left(\frac{\mu_B B}{k_B T}\right).$$

Substituting this, we find the cooling power:

$$Q = N_A \mu_B B_f \tanh\left(\frac{\mu_B B_f}{k_B T_f}\right).$$

where the final field B_f is the same as the interaction field b in this case. We have replaced the number of particles N by the

Avogadro constant N_A since we are given that there is one mole of the salt.

Substituting the above values for b and T_f and the constants, we find the cooling power:

$$Q = 0.0145 \text{ J}.$$

Next, we need to find the heat of magnetisation which has to be removed if this salt is magnetised isothermally to 2 T, at a temperature of 1 K. This can be calculated using the entropy:

$$Q = T\Delta S.$$

From the lectures on paramagnetic salts, we have seen that the entropy for the spin-1/2 salt is

$$S = Nk_B \ln\left[2\cosh\left(\frac{\mu_B B}{k_B T}\right)\right] - \frac{N\mu_B B}{T}\tanh\left(\frac{\mu_B B}{k_B T}\right).$$

where N should be N_A for one mole of the salt.

The entropy change refers to the change from the entropy at 1 K, 2T, to the maximum entropy of $N_A k_B \ln 2$.

Substituting 1 K, 2T into the entropy formula, we find

$$S = 1.612 \text{ J/mol}.$$

The entropy change ΔS is obtained by subtracting this from the maximum entropy of $N_A k_B \ln 2$.

We can now find the heat of magnetisation:

$$Q = T\Delta S = T(N_A k_B \ln 2 - 1.612) = 4.15 \text{ J}.$$

Exercise 3

To which temperature does one have to refrigerate a solid containing paramagnetic ions with spin 1/2 and magnetic moments equal to one Bohr magneton in a final field of 3 T so that 75% of the atoms are polarised with their spin parallel to the external magnetic field.

A spin 1/2 salt would have 2 magnetic energy levels: $-\mu_B B$ and $+\mu_B B$.

The Boltzmann factors for the 2 levels are respectively:

$$\exp\left(\frac{\mu_B B}{k_B T}\right) \text{ and } \exp\left(-\frac{\mu_B B}{k_B T}\right).$$

We need 75% of the atoms in the lower level, and 25% in the higher level.

So the ratio of the Boltzmann factor would be:

$$\exp\left(\frac{\mu_B B}{k_B T}\right) : \exp\left(-\frac{\mu_B B}{k_B T}\right) = 75 : 25,$$

or

$$\exp\left(\frac{2\mu_B B}{k_B T}\right) = 3.$$

We can solve this

$$\exp\left(\frac{2\mu_B B}{k_B T}\right) = 3.$$

for the temperature T. Rearranging, we get:

$$T = \frac{2\mu_B B}{k_B \ln 3}.$$

Substituting the value for B of 3 T and the other constants, we find

$$T = 3.67 \text{ K}.$$

Exercise 4

At which nuclear spin temperature T_n would copper nuclei order magnetically, if we assume that this order occurs roughly when the nuclear magnetic interaction energy μb_i becomes comparable to the thermal energy $k_B T_n$? The internal field created by the neighbouring nuclei $b_i \approx 0.3$ mT.

[You are given that copper has a nuclear g-factor of 2.22. The nuclear magneton $\mu_n = 5.051 \times 10^{-27}$ J T^{-1}.]

When the nuclear magnetic interaction energy μb_i becomes comparable to the thermal energy $k_B T_n$, we have

$$k_B T_n = \mu b_i.$$

The magnetic moment is

$$\mu = g\mu_n.$$

Substituting this, we get

$$k_B T_n = g\mu_n b_i.$$

Solving for T_n,

$$T_n = \frac{g\mu_n b_i}{k_B}.$$

Substituting the given values of g and b_i, we get

$$T_n = 3.66 \ \mu\text{T}.$$

Exercise 5

How long would it take a ^{3}He-^{4}He dilution refrigerator with a circulation rate of 1 mmole ^{3}He/s to precool 20 mole of copper in 8 T to 15 mK from a higher temperature? How does this compare with cooling it isothermally at 15 mK with the same dilution refrigeraton?

You may use the high temperature approximation for the entropy per mole:

$$S_n = R\ln(2I+1) - \frac{\lambda_n B^2}{2\mu_0 T_n^2}$$

where $\lambda_n = N_A I(I+1)\mu_0\mu_n^2 g_n^2/3k_B$, and g_n is the g-factor for the nucleus.

[You are given that copper has a nuclear spin I of 3/2 and a nuclear g-factor of 2.22.
The nuclear magneton $\mu_n = 5.051 \times 10^{-27}$ J T^{-1}.]

The high temperature approximation for the entropy is given by

In order to use this, x, given by

$$x = \frac{g_n \mu_n B}{k_B T},$$

must be small. Substituting the given B of 8 T, T of 0.015 K, and g_n of 2.22, as well as the other constants, we find

$$x = 0.43.$$

This is not much smaller than one, so using the high temperature approximation would only give us an estimate. But it should simplify the calculation a lot.

The cooling power of the dilution refrigerator is

$$\dot{Q} = 84\dot{n_3}T^2.$$

The flow rate $\dot{n_3}$ is rate of 1 mmole ^{3}He/s that is given. As the copper block is cooled from a higher temperature.

We also need to assume that the magnetic heat capacity is the main contribution to the heat capacity. We know from earlier lectures that the magnetic contribution becomes dominant at around this temperature range.

The heat absorbed from the copper is related to the entropy by

$$dQ = TdS.$$

This can be related to the cooling power as follows:

$$-\frac{dQ}{dt} = T\frac{dS}{dt}$$

A minus sign is introduced because absorbing heat from the copper causes its energy to decrease.

Substituting the above formula for the cooling power on the left, and the given formula for the entropy on the right, we get:

$$-84\dot{n_3}T^2 = \frac{n\lambda_n B^2}{\mu_0 T^2}\frac{dT}{dt}.$$

A factor n has been added for the given number of moles, since entropy formula is for 1 mole of copper. The derivative of T appears on the right because temperature changes with time. Rearranging gives:

$$\int_0^{t_1} dt = -\frac{n\lambda_n B^2}{84\mu_0\dot{n_3}}\int_\infty^{T_i}\frac{dT}{T^4}.$$

We use T_i to represent the temperature of 15 mK that we want to reach, and t_1 is the time taken.

We use infinity to approximate a high starting temperature. Since the integrating varies as $1/T^4$, it would become very small as high T, so this should be all right.

Integrating, we find the time taken:

$$t_1 = \frac{1}{3}\frac{n\lambda_n B^2}{84\mu_0\dot{n_3}T_i^3}.$$

Substituting the values:

$$n = 20,$$
$$B = 8 \text{ T},$$
$$T_i = 0.015 \text{ K and}$$
$$\dot{n}_3 = 0.001 \text{ mole/s},$$

we find the time taken:

$$t_1 = 10300 \text{ s}.$$

This is 2 hours and 52 minutes - quite fast in low temperature cooling.

Next, we need to compare this with isothermal cooling. Instead of starting at a higher temperature and cool down at a fixed field, we start at the low temperature and increase the field.

This is much easier to calculate. Since the temperature is constant the cooling power

$$\dot{Q} = 84\dot{n}_3 T_i^2$$

remains fixed. The heat of magnetisation can be calculated using TdS. Since temperature is constant, the heat change is

$$\Delta Q = T\Delta S.$$

The entropy change can be calculate from the the high temperature approximation for the entropy per mole:

$$S_n = R\ln(2I+1) - \frac{\lambda_n B^2}{2\mu_0 T_n^2}.$$

At zero field ($B = 0$ T),

$$S_n = R\ln(2I+1).$$

So the entropy change is obtained by subtracting the 2 equations above:

$$\Delta S = n\frac{\lambda_n B^2}{2\mu_0 T_i^2}.$$

where n is the number of moles of copper. The heat change is

then

$$\Delta Q = T\Delta S = \frac{n\lambda_n B^2}{2\mu_0 T_i}.$$

Let the time taken for the magnetisation be t_2. This time would depend on how fast the heat produced can be removed, i.e. the cooling rate $\dot{Q}$ of the dilution refrigerator. So the time taken is:

$$t_2 = \frac{\Delta Q}{\dot{Q}}.$$

Substituting the above equations, we find:

$$t_2 = \frac{1}{2}\frac{n\lambda_n B^2}{84\mu_0 \dot{n}_3 T_i^3}.$$

Comparing with the formula for t_1 above, we find that this time is longer, by 50%.

So it is faster in this case to cool down from a higher temperature!